

U.S. DEPARTMENT OF COMMERCE
National Technical Information Service

PB-298 548

Plutonium Uptake by Plants from Soil
Containing Plutonium-238
Dioxide Particles

(U.S.) Environmental Monitoring and Support Lab, Las Vegas, NV

20 Jan 78

TECHNICAL REPORT DATA (Please read Instructions on the reverse before completing)		
1. REPORT NO. EPA-600/J-78-115	2. JOURNAL ARTICLE	3. PB 298548
4. TITLE AND SUBTITLE PLUTONIUM UPTAKE BY PLANTS FROM SOIL CONTAINING PLUTONIUM-238 DIOXIDE PARTICLES	5. REPORT DATE	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) K. W. Brown and J. C. McFarlane	8. PERFORMING ORGANIZATION REPORT NO.	9. PERFORMING ORGANIZATION NAME AND ADDRESS Environmental Monitoring and Support Laboratory Office of Research and Development U.S. Environmental Protection Agency Las Vegas, NV 89114
10. PROGRAM ELEMENT NO.	11. CONTRACT/GRANT NO.	12. SPONSORING AGENCY NAME AND ADDRESS U.S. Environmental Protection Agency-Las Vegas, NV Office of Research and Development Environmental Monitoring and Support Laboratory Las Vegas, NV 89114
13. TYPE OF REPORT AND PERIOD COVERED Final	14. SPONSORING AGENCY CODE EPA/600/07	15. SUPPLEMENTARY NOTES Published in Health Physics, Vol 1 35 (September 1978), pp. 481-485. Contact: Kenneth W. Brown, FTS-595-2969, ext. 394 (Las Vegas, NV)
16. ABSTRACT Three plant species--alfalfa, lettuce, and radishes--were grown in soils contaminated with plutonium-238 dioxide ($^{238}\text{PuO}_2$) at concentrations of 23, 69, 92, and 342 rancuries per gram (nCi/g). The length of exposure varied from 60 days for the lettuce and radishes to 358 days for the alfalfa. The magnitude of plutonium incorporation as indicated by the discrimination ratios for these species, after being exposed to the relatively insoluble PuO_2 , was similar to previously reported data using different chemical forms of plutonium. Evidence indicates that the predominant factor in plutonium uptake by plants may involve the chelation of plutonium contained in the soils by the action of compounds such as citric acid and/or other similar chelating agents released from the plant roots.		
17. KEY WORDS AND DOCUMENT ANALYSIS		
a. DESCRIPTORS Plutonium-238 Metabolism Plant Growth	b. IDENTIFIERS/OPEN ENDED TERMS Uptake from soil	c. COSATI Field/Group 07B 066
18. DISTRIBUTION STATEMENT RELEASE TO PUBLIC	19. SECURITY CLASS (This Report) UNCLASSIFIED 20. SECURITY CLASS (This page) UNCLASSIFIED	21. NO. OF PAGES 8 22. PRICE A02-A01

NOTES

Plutonium Uptake by Plants Grown in Soil Containing Plutonium-238 Dioxide Particles

(Received 25 May 1977; accepted 20 January 1978)

Introduction

THE USE and production of transuranic elements by the expanding nuclear power industry, along with the release of many of these elements into the biosphere during periods of atmospheric nuclear testing, have prompted the need for assessing their behavior in biological systems. Of the transuranic elements being utilized and produced in these energy- and weapons-related programs, many investigators, including (Sa73) and (Fras67) consider plutonium, because of its half-life, to be one of the most radiologically and biologically toxic radioelements.

The environmental dissolution of plutonium particulates, which may become accidentally incorporated into man's metabolic system either by inhalation or ingestion, necessitates determining the retention and absorption characteristics of plutonium from sites of deposition.

Available literature identifying both the physical and biological parameters for assessing the amount of plutonium assimilated by plants via a soil rooting media is limited. In most controlled studies, plutonium is uniformly mixed in soil. As a result, the data from these experiments would not necessarily be correlatable to data collected from native vegetation growing in soils where the plutonium is deposited on the soil surface. Investigations by (Ro70) have indicated that plutonium is normally quite immobile and tends to remain in the upper few centimeters of the soil; therefore, it is not readily available for plant uptake. Nevertheless, the results of laboratory investigations where plutonium is uniformly mixed in the rooting media are valuable in understanding the contamination of cultivated vegetation grown on plowed lands and in identifying the mechanisms which control its uptake and distribution in plants. As such this investigation was designed to determine the extent and magnitude of plutonium dioxide assimilation by plants growing in premixed soils.

Methods and Materials

The chemical form and the isotope of plutonium selected for this study was $^{238}\text{PuO}_2$ particles. These monodisperse $^{238}\text{PuO}_2$ particles were obtained from the Inhalation Toxicology Research Institute, Lovelace Foundation, located in Albuquerque, New Mexico. The particles had a geometric mean diameter of 0.32 micrometers (μm) and were stored dry on stainless steel foil. The amount obtained for this study consisted of 2.6 mCi of ^{238}Pu with a trace amount of ytterbium-169 (850 nCi). The specific alpha activity of these particles was 13.6 Ci/g. The chemical composition by mass was 97% PuO_2 and 3% ytterbium trioxide ($^{169}\text{Yb}_2\text{O}_3$). The plutonium contained 90% ^{238}Pu and 10% ^{239}Pu by mass.

The soil selected for the rooting media was a silty loam consisting of 57.6% sand, 36.8% silt and 5.6% clay. It had a pH of 7.9 and a cation exchange capacity of 12.23 milliequivalents (m-equiv)/100 g.

The initial procedure for preparing the rooting media was to remove the particles from the foil and suspend them in a suitable solution. This procedure was previously described by (Ra75). Basically this method involves adding 50 ml of a 0.02% surfactant solution (Triton $^{\circledR}$ X-100) to the centrifuge tube, thereby submerging the stainless steel foil. The centrifuge tube was then placed into an ultrasonic water bath to dislodge the plutonium particles from the stainless steel foil. After a 4-hr period of ultrasonic agitation, the foil was removed.

Because of the necessity of dry mixing to obtain a homogeneously mixed rooting media, aliquots of the surfactant solution containing the plutonium particles were added to a slurry of talc, $\text{H}_2\text{Mg}_3(\text{SiO}_3)_4$. The talc, which readily absorbed the liquid, was then dried under an i.r. lamp. The talc mix, which dried into a brittle conglomerate, was transferred *in toto* to a 30-l. capacity Patterson-Kelley Twin Shell $^{\circledR}$ blender for mixing.

The homogeneity of the soil-plutonium mix was determined over a 20-hr blending period by taking soil samples from the blender at various times and analyzing for the ^{169}Yb content using a gamma scintillation detector. Four different soil-

plutonium concentrations (mixes) were made for this study, each consisting of approx. 5100 g. After mixing, each of the four soil mixes was divided into six nearly equal portions and put into 1000-g vol. plastic bottles.

The ^{238}Pu contaminated soil was transferred from the plastic bottles into specially designed 12 cm greenhouse pots. Before transferring the soil, 25 g of vermiculite was added to each of the 24 plastic bottles to prevent excessive soil compaction during plant growth. The bottles, which contained approx. 850 g of the plutonium-contaminated soil, were capped and then rotated by hand for approx. 5 min to mix the vermiculite into the soil. After mixing, all the soil from one of the bottles was poured into one of the 12 cm pots. This procedure was duplicated until all 24 pots were filled. The pots were then placed into a self-contained environmental growth chamber.

The pots, as shown in Fig. 1, were designed to contain the plutonium over an extended period. To prevent loss of the plutonium particles by leaching, a nylon reinforced Acropor[®] filter with a pore size of $0.20\text{ }\mu\text{m}$ was cemented over the drain holes. To protect the Acropor filter from damage by roots and to prevent it from being plugged by soil particles, a Microsorban[®] filter was placed in the bottom of the pot to act as a prefilter. To reduce the loss of the plutonium particles by upward migration via capillary action, the soil surface was covered by a Whatman[®] filter that had been impregnated with seeds. The Whatman filter was then covered with a 3.0-cm deep layer of 0.3-cm dia polystyrene beads.

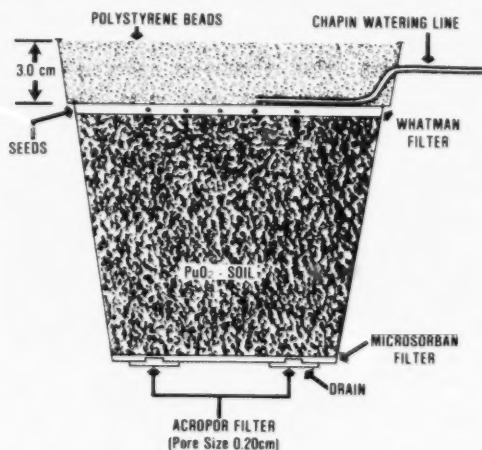


FIG. 1. Greenhouse pots used to hold the $^{238}\text{PuO}_2$ -contaminated soil.

Further safety precautions included the construction of a fiberglass-lined wooden tank measuring $115 \times 115 \times 18\text{ cm}$ which was placed in the growth chamber to hold the pots. Once the pots were placed in the tank, handling of the contaminated material was eliminated except during harvesting. Also, an automatic irrigation system was designed. Features of this irrigation system included the recycling of evapotranspired water, exterior controls, and a safety float installed in the fiberglass-lined tank. To ensure a fairly even distribution of water to each pot, four manifolds were used, each distributing water to six pots. Plant nutrients were provided by irrigating once every six weeks with a modified Hoagland solution as described by (Berr71).

An air sampling system was constructed to sample the chamber air. The air was pumped from the chamber through a Millipore[®] membrane filter having a pore size of $0.1\text{ }\mu\text{m}$ and then exhausted back in the chamber. The chamber air was sampled for approx. 6 hr at a flow rate of 12 liters per minute (l/min). This sampling was conducted 24–36 hr prior to entering the chamber. After the decay of naturally occurring radon, the filter was analyzed to determine if any of the plutonium-238 had become airborne.

Throughout this investigation a photo-period of 16 hr was maintained. The light-dark temperatures were kept at 25°C and 20°C , respectively. Carbon dioxide (CO_2) was automatically injected into the chamber atmosphere to maintain a uniform day-time concentration of 350 parts per million (ppm).

Samples and sample analysis

Soil samples were collected from each pot 2 days after the pots were placed in the chamber. This 2-day delay was to ensure that all of the soil in the pots was damp due to irrigation. Samples were collected by inserting a 10-cc disposable syringe, which had the bottom (needle attachment) end cut off, into the soil. The syringe was then withdrawn containing a 7- to 10-g (dry weight) core of soil. To avoid cross-contamination, 24 different syringes were used. The wet weight of each soil sample was determined and then they were dried in an oven at a temperature of 100°C to determine the dry weight.

Plant samples were collected by clipping with scissors and then placed into small preweighed paper bags. The samples were weighed and then dried at a temperature of 75°C . The bags were individually sealed in aluminum cans and along with the soil samples sent to the Eberline Instrument Corporation of Albuquerque, New Mexico for ^{238}Pu analysis. The plant samples were not

removed from the paper bags but were dissolved *in toto*. This procedure eliminated additional handling of plant material and therefore increased the precision of analysis. No attempt was made to separate the various plant organs, for example, stems from leaves. Following a period of regrowth, usually from 5 to 6 weeks, the plants were again reharvested.

Basically, the analytical technique included decomposition of the soil and plant material by potassium fluoride fusion and/or acid dissolution. After decomposition, ^{239}Pu was added as a tracer followed by the separation of the plutonium by ion exchange or solvent extraction. The plutonium was electroplated and then counted by alpha spectroscopy.

Results and Discussion

Two plant species, alfalfa, *Medicago sativa*, and lettuce, *Lactuca sativa*, were originally planted. Three replications of each of the four plutonium-soil mixes, were sown with each species. All of the alfalfa germinated; however, only 3 of the 12 pots of lettuce germinated. As a result, after harvesting the lettuce, all 12 of the pots originally planted with this species were resown with radish, *Raphanus sativus*.

The analytical results of the four soil mixes were 23 ± 3 , 69 ± 8 , 92 ± 3 and 342 ± 10 nCi/g. These results are also shown in Table 1. In soil treatments 1 and 2, the coefficient of variation were 13 and 12%, and only 3% in treatments 3 and 4. These soil concentration values compare closely with the soil concentrations calculated and analyzed by gamma counting the ^{169}Yb . The results based on the ^{169}Yb concentration for the four treatments were 24, 62, 94 and 310 nCi/g, respectively. The use of the ^{169}Yb as an analytical tool for determining the ^{238}Pu concentration in the treatment 1 and 2 soils indicated that much less variation existed in each of these two mixes when compared to the soil ^{238}Pu analyses, as the standard deviations were calculated to be only 7 and 6% of the means. It is assumed that the larger variability (13–12%) was the result of the ^{238}Pu analysis rather than in the soil preparation techniques. The variation in the treatment 3 and 4 soils was nearly identical after the two analyses.

Investigations involving the transfer of plutonium from soils to plants via root assimilation have generally shown that a concentration ratio (CR)

(CR) =

$$\frac{\text{plutonium concentration in the plant (nCi/g dry)}}{\text{plutonium concentration in the soil (nCi/g dry)}}$$

of approx. 10^{-5} exist. The concentration ratios calculated from this study are presented in Table 1 for each of the three species.

The magnitude of plutonium uptake appeared to be greater in both the lettuce and radishes than it did in the alfalfa as indicated by the concentration ratios shown in Table 1. However, the ratios calculated for the lettuce and radishes are in most cases based on a single harvest and are considered to be inconclusive as an indication of species differences or trends in plutonium incorporation.

Francis (Fran73) summarized the available literature and reported that the concentration ratio generally falls between 10^{-4} and 10^{-6} . The alfalfa concentration ratios shown in Table 1 are similar in magnitude to those previously reported. It is somewhat surprising to find that these values, which represent the absorption and translocation of plutonium from soils contaminated with discrete spheres of relatively insoluble PuO_2 , are similar to those in which ionic and chelate-complexed plutonium were applied as the contaminant. Even experiments reported by (McF76), in which plant roots were treated with plutonium in solution cultures, showed discrimination ratios against plutonium in the same general magnitude. This suggests that the chemical reactions which occur at the root surface predominate the kinetics of plutonium uptake and translocation in plants. Plants are known to exude organic compounds such as citric and humic acids which form strong chelates with plutonium. Based on the results of this study and the results from other investigations (Ro70), it seems possible that the release of citric acid and/or other similar chelating compounds may be responsible for plutonium uptake.

Table 1. Plutonium concentration ratios for alfalfa, lettuce and radish plants grown in soils contaminated with $^{238}\text{PuO}_2$ spheres

Treatment	Soil concentration (nCi/g)	Concentration ratios		
		Alfalfa	Lettuce	Radish
1	$23 \pm 3^*$	$7.4 \pm 3.0^* \times 10^{-5}$	2.6×10^{-5}	3.4×10^{-4}
2	69 ± 8	$7.5 \pm 4.0 \times 10^{-5}$	1.7×10^{-4}	1.2×10^{-4}
3	92 ± 3	$8.8 \pm 4.0 \times 10^{-5}$	1.8×10^{-4}	4.2×10^{-5}
4	342 ± 10	$7.4 \pm 3.8 \times 10^{-5}$		2.7×10^{-4}

*Standard deviations, s, of six soil analyses.

†Standard deviation, σ , of ten concentration ratios.

If the absorption of plutonium by plants is dependent on the release of a particular compound or on the formation of some chemical complex at the root surface, it would explain why plutonium solubility in soil, water movement in plants, growth rate, and root contact potential appear to have little impact on plant assimilation of plutonium. This would also explain the tight grouping of discrimination ratios for dissimilar experiments where the chemistry of the contaminating plutonium was extremely different.

The growth rate (dry matter synthesis) of the alfalfa in this study increased with each successive cutting. This increase is evidence that the rooting systems were increasing in size, therefore, coming in physical contact with more potentially absorbable plutonium. A proportionate increase in total plutonium uptake and translocation was associated within this increased plant growth rate. However, the concentration in the plant tissue remained unchanged and was apparently independent of time or root exposure. At one point in the experiment the growth chamber overheated due to a mechanical failure. Severe wilting occurred followed by the harvesting of the alfalfa stems and leaves. Subsequent growth was suppressed and damage to the root system was suspected. Despite this stress, no detectable change occurred in the rate of plutonium uptake, the plant concentration, nor in the concentration ratio.

For a number of reasons, a relatively few laboratory plant kinetics studies involving plutonium are conducted over an extended period of time. However, one study conducted and reported by Dr. Romney and his research associates in 1970, was in many aspects similar to this investigation. In their study, Nevada Test Site (NTS) soils contaminated with various chemical forms of plutonium were used as the rooting media. Close similarities between the two investigations were that both were cropping studies conducted over a relatively long time, and they both utilized plant species belonging to the Leguminosae family.

Dr. Romney *et al.* (Ro70) reported that the Ladino clover that they grew and cropped over an extended 5-yr period had plutonium concentration ratios that varied from 1.9×10^{-5} to 14.0×10^{-5} . They also reported that a trend of increasing plutonium uptake by this plant species occurred with time. Their data support the hypothesis that plutonium incorporation by plants may be primarily dependent upon the release of chemical compounds from plant roots and/or from microbial action. Even though the magnitude of plutonium incorporation by the alfalfa was similar to that taken up by the Ladino clover as shown by the

concentration ratios, no similar increase of plutonium uptake by the alfalfa as a function of time was evident. The absence of this trend may have been due to the comparatively short duration of the alfalfa exposure which would reduce the amount of soil chemical formation.

Conclusions

The results of this study indicate that the magnitude of assimilation and translocation of $^{238}\text{PuO}_2$ by these plants is in about the same proportion as the incorporation of other chemical forms of plutonium by a variety of other plants, including both aquatic and terrestrial species. The long-term exposure of the alfalfa did not show any increase in the concentration of plutonium in the plant tissue, even though the root mass increased. This increase in root mass would normally enhance the probability of a contaminant assimilation as the change of physical contact with the soil-borne pollutants would increase.

As the behavior of $^{238}\text{PuO}_2$ in soils, as indicated in this study, parallels other chemical forms of plutonium as far as plant assimilation, the rate and means of uptake may be largely determined by the effect of root exudates. A number of investigators, including (Ro70) and (Pr72), have indicated that the biological availability of plutonium is largely governed by its solubility and also to the numerous chemical reactions which occur in soils. These reactions, which are enhanced by the soil microflora, as shown by (Au74), involve and affect soil pH and the rates of natural and/or induced chelation. As such, the effects of chelation, additions of various soil dressings and a changing soil pH on the availability of plutonium transfer from soils to plants are important factors that merit further study.

Acknowledgements—The authors would like to express their appreciation to Dr. O. G. Raabe and Dr. R. O. McClellan of the Inhalation Toxicology Research Institute, Lovelace Foundation, for providing the plutonium-238 dioxide microspheres used in this study.

K. W. BROWN

J. C. MCFARLANE

Environmental Monitoring and Support Laboratory

U.S. Environmental Protection Agency

P.O. Box 15027

Las Vegas, NV 89114

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0017-9078/78/0901-0485/\$02.00/0

Health Physics Vol. 35 (September), pp. 485-489
 Pergamon Press Ltd., 1978. Printed in Great Britain
 Health Physics Society

Leakage Radiation Characteristics of an 18 MeV Clinical Linear Accelerator*

(Received 1 January 1977; accepted 17 February 1978)

Introduction

AN INCREASING number of high energy linear accelerators for radiation treatment with X-rays and electrons currently are being installed in this country. One of the first concerns of the purchaser is that the unit can be used safely with regard both for the patient and for the operators. Part of a routine investigation into the safety aspects of the use of this type of unit is an evaluation of the leakage radiation characteristics (As67; Ca68; Ka67; La51; Le70; Ma59; Wi72).

We have recently commissioned a linear accelerator which is of a relatively new design. This unit produces an X-ray beam of 10 MV as well as electron beams of 6, 9, 12, 15 and 18 MeV. While the treatment beams from this linear accelerator may vary in energy somewhat from one machine to the next, some of the leakage radiation characteristics which we describe here should be of interest to other users as well as to prospective buyers of this type of machine.

Measurements

Film detection

The spatial distribution of leakage radiation was determined using pre-packaged 14×17 in Kodak Industrial Type M film. No attempt was made to evaluate the densities obtained as described by Capone and Karzmark (Ca68) because of the energy dependent response of film and the likelihood of finding areas of high film density caused by low energy, multiple scattered X-rays and/or electrons.

Films were exposed in groups of from three to

*Work supported in part by NIH grants CA-14520, CA-05104 and CA-16127 from the National Cancer Institute.

seven films taped directly to the treatment head shielding and support structures. Several separate groups of films were used to cover the entire treatment head. An attempt was made to obtain similar densities on the films as each group of films was at a somewhat different distance from the target of the accelerator and therefore required somewhat different exposures.

The bottom area of the gantry proximal to the collimator assembly was covered with three films in Group 1. Film Group number 2 consisted of five films taped together to form a cylinder around the collimator assembly. The seven films in Group 3 were placed around the end of the treatment head overlapping the position of Group 2 films by about 3 in and extending well back toward the electron gun portion of the accelerator. The seven films in Group 4 overlapped the position of the Group 3 films by about 4 in. The top of the gantry was covered by three films in Group 5.

The position of these groups of films are illustrated in Fig. 1. Areas of significant leakage radiation are shown with the number indicating the film and the lower case letter to indicate a particular

area on the film. The radiation levels at these positions were evaluated using an ionization chamber technique described in the following section.

Ionization Measurements

Leakage radiation levels were measured using a Victoreen 666 electrometer and "background probe". The Victoreen "background probe" is an ionization chamber with a volume of 257 cm³ and a wall thickness of 300 mg/cm².

Because of the relatively thin wall of the ionization chamber used, all measurements were made with an uncovered chamber and repeated with the chamber covered with a thin lead sheet build-up cap. The ratio obtained in this way gives an approximate indication of the mean photon energy being detected. This is an important factor when the radiation field is made up largely of scattered photons and electrons.

Ionization measurements were made at the 14 primary points described in NCRP Report No. 33 (NCRP 68) for cobalt teletherapy units as well as those points indicated by the films described in the previous section.

Figure 2 shows the position of the ion chamber measurements made at the 14 primary points surrounding the treatment head. The results of these measurements are shown in Table 1.

In Table 2 the results of the ionization chamber

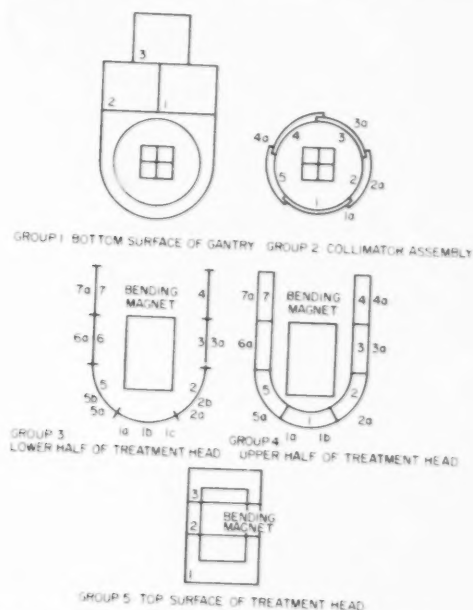


FIG. 1. Positions of film groups covering the treatment head to determine the spatial distribution of leakage radiation. Each film is numbered. Each area of significant radiation is indicated by a lower case letter as well as the film number.

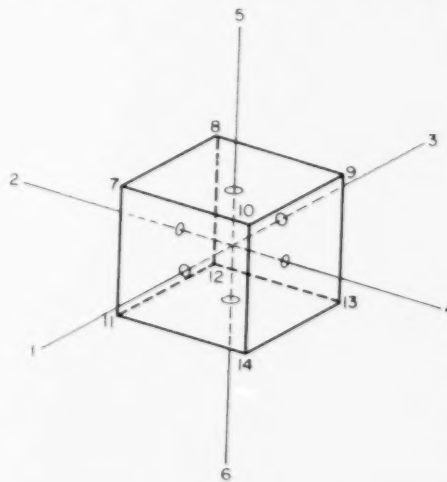


FIG. 2. Diagram illustrating the 14 primary points of measurement around the treatment head. The target is located at the intersection of lines 1-3, 2-4 and 5-6.